

Comment on “Hole digging in ensembles of tunneling molecular magnets”

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Tupitsyn et al. [Phys. Rev. B **69**, 132406 (2004)] have recently reported results for the relaxation of crystalline systems of single-molecule magnets, such as Fe₈. They claim that, quite generally, (1) the magnetization and hole widths of field-distributions evolve with time t as \sqrt{t} , and (2) the holes' line shapes are Lorentzian. We give a counter-example to these conclusions, and show that the main assumption on which some of them rest is invalid.

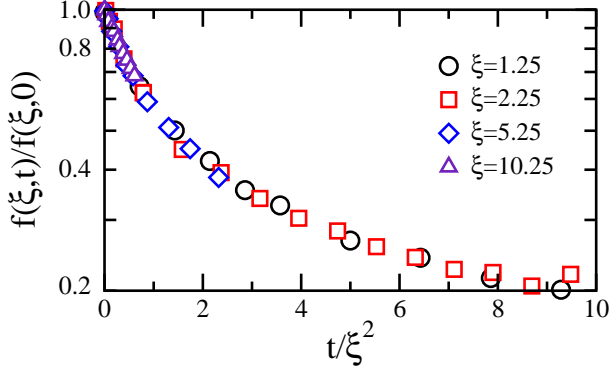


FIG. 1: The field function $f(\xi, t)$ versus t/ξ^2 for SC lattices and the shown values of the dipole field ξ . The tunnel window's half-width ξ_0 is 0.1. The initial value of the magnetization m_0 is 0.2 of saturation. All data points are for systems of 32768 spins, and follow from averages over 800 runs, over times much greater than $t \gg \tau_0$ but within the range where $m \propto t^p$.

Tupitsyn, Stamp, and Prokof'ev¹ (TSP) have recently reported results for the relaxation of crystalline systems of single-molecule magnets, such as Fe₈, whose tunneling windows are much narrower than their dipolar field spread. They find that, independently of crystalline lattice structure, (1) the magnetization m and hole widths of field-distributions evolve with time t as \sqrt{t} , and (2) field-distribution line shapes are Lorentzian. The first conclusion is contrary to our prediction, of Ref. [2], that, in zero field, m relaxes from weakly polarized states as t^p , where p depends on crystal structure. We give below a counter-example to the conclusions TSP have reached, and show that the main assumption on which some of their conclusions rest is incorrect.

We mainly use the notation of Ref. [1], giving the bias field ξ in terms of the tunnel window field ξ_0 , but ξ_0 is given in terms of the nearest neighbor dipolar field E_D . We assume spins flip at rate $1/\tau_0$ if $|\xi| < 1$, but not at all otherwise, and time t is given in terms of τ_0 . The following numbers may be found useful: the rms value of the dipolar field $\delta\xi$ is $3.7E_D$ and $8.3E_D$ for SC and FCC, respectively.

Let $p_\uparrow(\xi, t)$ [$p_\downarrow(\xi, t)$] be the number density of up-spins (down-spins) with a field ξ acting on them, and let $f(\xi, t) = p_\downarrow(\xi, t) - p_\uparrow(\xi, t)$. Note that $m(t) =$

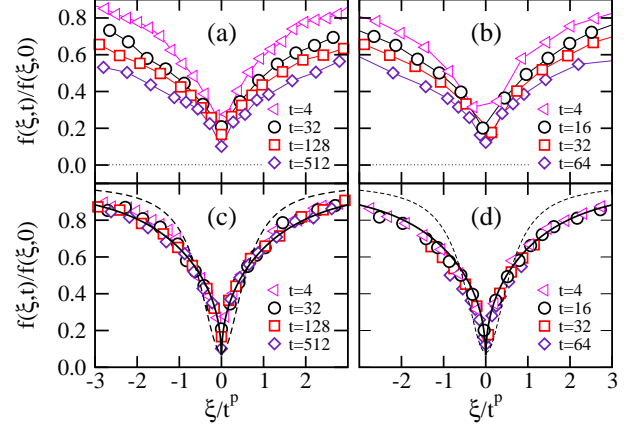


FIG. 2: (a) $f(\xi, t)$ versus ξ/t^p for FCC lattices and the shown values of t , $\xi_0 = 0.01$, and $p = 0.5$. Lines are guides to the eye. (b) Same as in (a) but for $\xi_0 = 0.1$. (c) Same as in (a) but for $p = 0.726$. The dashed line is for the best fitting Lorentz curve. The full line is from our theory. (d) Same as in (c) but for $\xi_0 = 0.1$. In all cases, $m_0 = 0.2$. All data points are for systems of 65536 spins, and follow from averages over 800 runs over times much greater than $t \gg \tau_0$ but within the range where $m \propto t^p$. All the shown data points come from the $2\xi_0 < \xi < 0.2\delta\xi$ (nearly one and two decades for $\xi_0 = 0.1$ and $\xi_0 = 0.01$, respectively). In the shown range of time values, $1 - m/m_0$ changes by approximately one and two decades (see Fig. 1), for $\xi_0 = 0.1$ and $\xi_0 = 0.01$, respectively.

$-\int d\xi f(\xi, t)$. The main ingredient underlying Eq. (1) of Ref. [1] is the assumption that $f(\xi, t) \propto N(\xi) \exp[-t/\tau(\xi)]$, where $N(\xi)$ is of no interest to us here, and $\tau(\xi)$ is some time that depends only on ξ . The Monte Carlo (MC) results shown in Fig. 1 are contrary to the assumption of TSP, that $f(\xi, t)$ is exponential in t .³ [The probability density that a spin have field ξ and has not yet flipped at time “ t ” behaves much as $f(\xi, t)$.]

From the assumption that $f(\xi, t) \propto \exp[-t/\tau(\xi)]$ and the further general statement TSP make, that $1/\tau(\xi)$ is a Lorentzian function of ξ , hole line widths that grow as \sqrt{t} when $t \gtrsim \tau_0$ follow in Ref. [1]. Such \sqrt{t} growth does take place in SC lattices, but not in general, as one can gather from the MC results exhibited in Figs. 2a and 2b for FCC lattices. Rather, from Figs. 2c and 2d, one gathers that ξ then scales as t^p , where $p \simeq 0.73$.⁴ Finally,

in the region of interest, when $t \gg \tau_0$ but t is still within the range where $m \propto t^p$, the line shape in Figs. 2c and 2d differ significantly from a Lorentzian function. For comparison, results from our own theory⁵ are also shown in Figs. 2c and 2d.

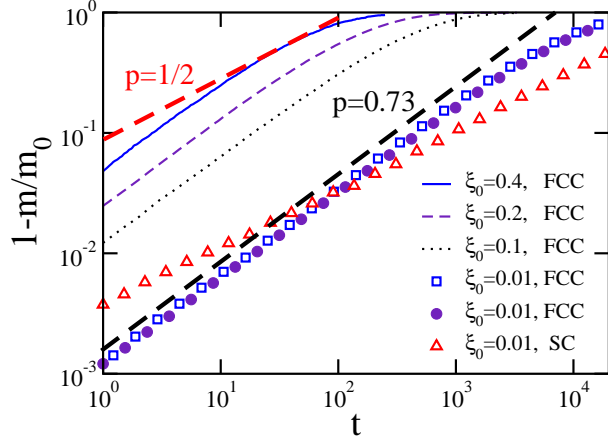


FIG. 3: $1 - m/m_0$ versus time t . All data points follow from MC simulations. Triangles are for SC lattices. All other data points are for FCC lattices. All data points for the FCC lattice are for systems of 8192 spins, except for \bullet , which stand for 65536 spins. Data points for SC lattices are for 4096 spins. For all data points, averages over 1200 runs were performed, except for the 65536 spin system and $t > 1200$. For them, averages over 100 runs were performed.

For completeness sake, we examine further numerical evidence that supports our claim that $p = 0.73$, not $1/2$, for FCC lattices. To this end, note first that $\xi \sim t^p$ scaling implies, through the relation $m(t) = -\int d\xi f(\xi, t)$, that $m \sim t^p$. Thus, the value of p can also be obtained from the time evolution of m .

The difference between the relaxation of the magnetization in SC and FCC lattices can be clearly appreciated in Fig. 3, as well as in Figs. 4a and 4b. Note in Fig. 3 that, for FCC lattices, $m \propto t^p$ and $p \simeq 0.73$ for time spans that are increasingly larger for larger values of $1/\xi_0$. (This is as predicted in Ref. [2].) Note also that the $p = 1/2$ slope that is claimed by TSP to hold universally for all lattices appears to ensue for FCC lattices only when the relaxation crosses over from the $m \propto t^p$ regime to saturation. This is in clear contrast with the data points for SC lattices in Fig. 3, for which $p \simeq 0.5$. Linear m/m_0 versus time plots for SC and FCC

lattices which further illustrate this point are also shown in Figs. 4a and 4b. The small neighborhood of the inflection point (marked as a straight line segment) in Fig. 4b is, of course, nearly straight. This transient behavior might be misinterpreted as the onset of a $m \propto \sqrt{t}$ regime if, as in Fig. 1 of Ref. [6], data points below $m/m_0 \simeq 0.5$ are not included in the plot.

Finally, comparison of the results shown in Figs. 2a and 2b, Figs. 2c and 2d, as well as among different curves shown in Fig. 3 for different values of ξ_0 should allay any

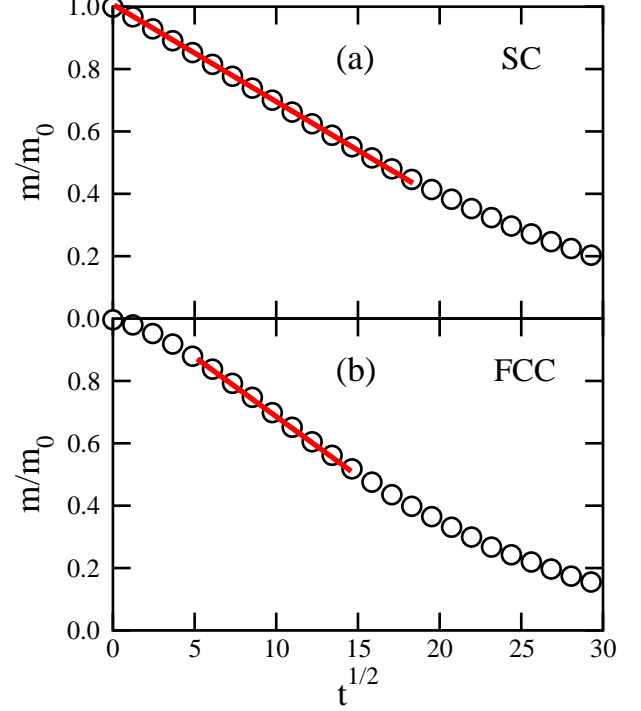


FIG. 4: (a) m/m_0 versus \sqrt{t} . Data points are from averaging over at least 4000 MC runs for 4096 spins on SC lattices with $\xi_0 = 0.1$. The straight line is a guide to the eye. (b) Same as in (a) but for 8192 spins on an FCC lattice. The straight line segment covers a neighborhood of the inflection point.

misgivings about spurious effects that might arise from the finite number of spins n_s in the tunnel window, since $n_s \propto \xi_0$ if $\xi_0 \ll \delta\xi$. To the same end, data points for two FCC lattice sizes are shown in Fig. 3 for $\xi_0 = 0.01$.

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¹ I. S. Tupitsyn, P. C. E. Stamp, and N. V. Prokof'ev, Phys. Rev. B **69**, 132406 (2004).

² J. F. Fernández and J. J. Alonso, Phys. Rev. B **69**, 024411 (2004); see also J. F. Fernández and J. J. Alonso, Phys. Rev.

Lett. **91**, 047202 (2003); I. S. Tupitsyn, P. C. E. Stamp, Phys. Rev. Lett. **92** 119701 (2004); J. F. Fernández and J. J. Alonso, Phys. Rev. Lett. **92** 119702 (2004).

³ This is because $\partial f(\xi, t)/\partial t \neq -f(h, t)/\tau(\xi)$, since $\partial f(\xi, t)/\partial t$ depends, not only on f and ξ , but also on the rate of magnetization reversal which is going on within the tunneling window at time t , which in turn depends non-

trivially on time.

⁴ This number is as given in our Phys. Rev. B paper in Ref. [2].

⁵ J. F. Fernández and J. J. Alonso, to be published somewhere

else.

⁶ I. S. Tupitsyn, P. C. E. Stamp, and N. V. Prokof'ev, arXiv:cond-mat/0407713 v1 27 Jul 2004.